

# Current status and future of transactinide heavy element chemistry

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## 1. Introduction

The knowledge and understanding of the chemistry of the superheavy elements, especially of the early transactinides Rf, Db and Sg and their compounds, both experimentally and theoretically, is nowadays quite extensive as documented by a first textbook entitled "The Chemistry of Superheavy Elements" edited by M. Schädel [1]. While rapid chemical separations in aqueous solution were (and are) the method of choice for detailed investigations of Rf and Db, the development of gas-phase chemical separation methods have allowed first chemical studies of increasingly heavier transactinide elements.

The spectacular results obtained at Flerov Laboratory in Dubna using  $^{48}\text{Ca}$  ion beams and targets of  $^{238}\text{U}$ ,  $^{242,244}\text{Pu}$ ,  $^{248}\text{Cm}$  and  $^{249}\text{Cf}$  to synthesize isotopes of elements 112 [2], 114 [3,4], 116 [5] and 118 [6] give chemists the perspective to experimentally investigate the chemistry of even the heaviest known elements. Especially since some of the isotopes attributed to elements 108, 110, 112, and 114 seem to have half-lives of the order of seconds to minutes.

## 2. Status

Since 1999 when the first TAN conference was held in Seeheim, Germany, chemists have made a quantum leap and are now working with nuclides that can be produced with picobarn ( $10^{-36} \text{ cm}^2$ ) cross sections only. One of the highlights of TAN'99 was the announcement of the first gas-phase chemical isolation of bohrium ( $Z=107$ ) as volatile  $\text{BhO}_3\text{Cl}$  using the OLGA technique [7]. The nuclides  $^{266}\text{Bh}$  and  $^{267}\text{Bh}$  were produced in the reaction  $^{249}\text{Bk}(^{22}\text{Ne}, 4,5n)$  with maximum cross sections of about 100 pb [8]. Even though the experiment was successful and deserved highest scientific merits, the outlook to apply the same technique to the investigation of even heavier elements was bleak, since the overall efficiency to detect a correlated mother-daughter  $\alpha$ -particle decay chain was only about 4%. The experimental chemical investigation and characterization of the next heavier transactinide element hassium has, for some years, constituted a daunting task even though from the very beginning the selection of a volatile compound was absolutely clear. Hassium, as a presumed member of group 8 of the Periodic Table and thus a homologue of Fe, Ru, and Os, should form stable and at the same time very volatile  $\text{HsO}_4$  molecules, very similar to  $\text{OsO}_4$ . However, in order to gain access to hassium ( $Z=108$ ) the overall efficiency of the experiment had to be improved by at least one order of magnitude. By developing a completely new approach of separation (IVO) [9] and detection (CTS, COLD) [10] and also by introducing the rotating target set-up ARTESIA [11], which allowed the use of much higher beam intensities, the first chemical characterization of Hs became reality [12]. This spectacular progress however is limited to a few selected cases which chemically are extremely favorable. One such case is also element 112 in its elemental state. First experiments made in Dubna [13,14] and recently at GSI [15] seem to indicate a quite spectacular difference in chemical behavior between element 112 and its lighter homolog Hg in the elemental state. Compared to chemical investigations of the early transactinide elements Rf through Hs, where always the highest possible oxidation state was investigated, now with elements 112 through 118,

which are expected to be moderately to highly volatile in the elemental state, the influence of relativistic effects of the valence electrons should become noticeable in direct experimental investigations. These relativistic effects might be responsible for some quite dramatic differences in chemical behavior as this seems to be the case in the adsorption properties of element 112 compared to its lighter homologue Hg [16].

Experiments on the aqueous chemistry of transactinide elements focused mainly on a much deeper understanding of the properties of Rf in solution [17]. A fully Automated Ion exchange apparatus coupled to a Detection system for Alpha-particle spectroscopy (AIDA) developed at JAERI, was used to extensively study the behavior of Rf in pure HCl solution [18]. Future experiments with Db using the reaction  $^{248}\text{Cm}(^{19}\text{F}, 5\text{n})^{262}\text{Db}$  are planned [18]. A series of trendsetting experiments were performed by the SISAK collaboration [19]. The nuclide  $^{257}\text{Rf}$  was produced in the reaction  $^{208}\text{Pb}(^{50}\text{Ti}, 1\text{n})^{257}\text{Rf}$ , separated in the Berkeley Gas-filled Separator (BGS) and transferred to a gas jet using the Recoil Transfer Chamber (RTC) [20]. The activity delivered by the gas jet was dissolved in nitric acid and Rf was extracted into dibutyl-phosphoric acid in toluene. The decay of Rf nuclides was registered on-line by flow-through liquid scintillation counters. This was the first time a transactinide element was extracted and unequivocally identified by the SISAK system. The use of the BGS provided the necessary reduction of background from interfering by-products of the nuclear reaction. Another approach to extend liquid-phase chemical studies to heavier transactinides is the so-called Multi Column Technique (MCT) [21]. In this continuous on-line chromatography, the solution containing the dissolved transactinide element passes through a series of columns. The transactinide element of interest passes a first column where only its actinide daughter nuclei (and other actinides) are retained. The transactinide element is then retained on a second column. However, its actinide decay daughters pass this column and are retained on a third column from which they are eluted after completion of the experiment and assayed by alpha-particle spectroscopy. This technique is very efficient and allows to access relatively short-lived nuclei. However, a breakthrough even of a small fraction of actinides in the first column can ruin the experiment. Also, all information about the nuclear decay properties of the transactinide element is lost.

Heavy element chemistry is not only dependent on outstanding radiochemists, but also on the necessary infrastructure consisting of powerful heavy ion accelerators and facilities and equipment to produce and handle highly radioactive and also extremely valuable exotic target materials such as  $^{248}\text{Cm}$ . In this respect the greatly reduced availability or even the closure of the 88-Inch cyclotron at LBNL without adequate replacement signifies a hard blow to our community. In this respect it is of importance to note that new groups have entered the field of transactinide chemistry, such as the group at JAERI. Remarkable is also the synthesis of the new nuclide  $^{259}\text{Db}$  at IMP Lanzhou, China [22] allowing also future transactinide chemistry experiments.

### 3. Future of transactinide chemistry

The spectacular results obtained at Flerov Laboratory in Dubna in the synthesis of new superheavy elements already have stimulated chemists to experimentally investigate the chemistry of even the heaviest known elements. A first step into this direction certainly are the chemical experiments performed with the spontaneously fissioning nuclide  $^{283}112$ . Nevertheless, it is highly desirable to develop techniques that are sensitive to reveal also isotopes that decay by chains of  $\alpha$ -particles in order to independently verify the results obtained by kinematical separator systems.

What are the prerequisites to achieve the ultimate goal of chemically identifying a new, superheavy element? Due to the very low production cross sections of 1 pb or less highest possible beam intensities must become available in the near future. This ultimately calls for a new accelerator capable of delivering a continuous beam of up to  $5 \cdot 10^{13}$  ions/s. Evidently, this also calls for improved target irradiation facilities,

since target thicknesses of the order of 1 to 1.5 mg/cm<sup>2</sup> are useful. Nevertheless, the experiences with ARTESIA clearly demonstrated, that this setup already today allows accepting a 3 to 4 fold higher beam intensity if a DC beam were available instead of a pulsed beam.

Chemical separation procedures need to be improved with respect to sensitivity and speed. Here, gas phase chemical separations will play an important role, since many of the superheavy elements with atomic numbers between 112- 118 may be moderately or even highly volatile in their elemental state. Thus future experiments point to investigations of elements 112 and 114 in their elemental state. However, a particularly difficult problem constitute some Rn isotopes, since their Po daughter nuclides decay with  $\alpha$ -decay energies which strongly interfere with the detection of heavy transactinide nuclides. Especially element 112 seems to exhibit a very similar volatility as Rn. A possible solution to this problem, although not yet available, may constitute a dedicated kinematical pre-separator which is coupled to chemical set-ups. A workshop held a GSI in 2002 clearly demonstrated the future need for such an instrument. Possible pathways to gas chemical investigations of element 112 and 114 are outlined in Fig.1. The currently most convenient production reaction of element 114 is the reaction  $^{48}\text{Ca}+^{244}\text{Pu}$ .

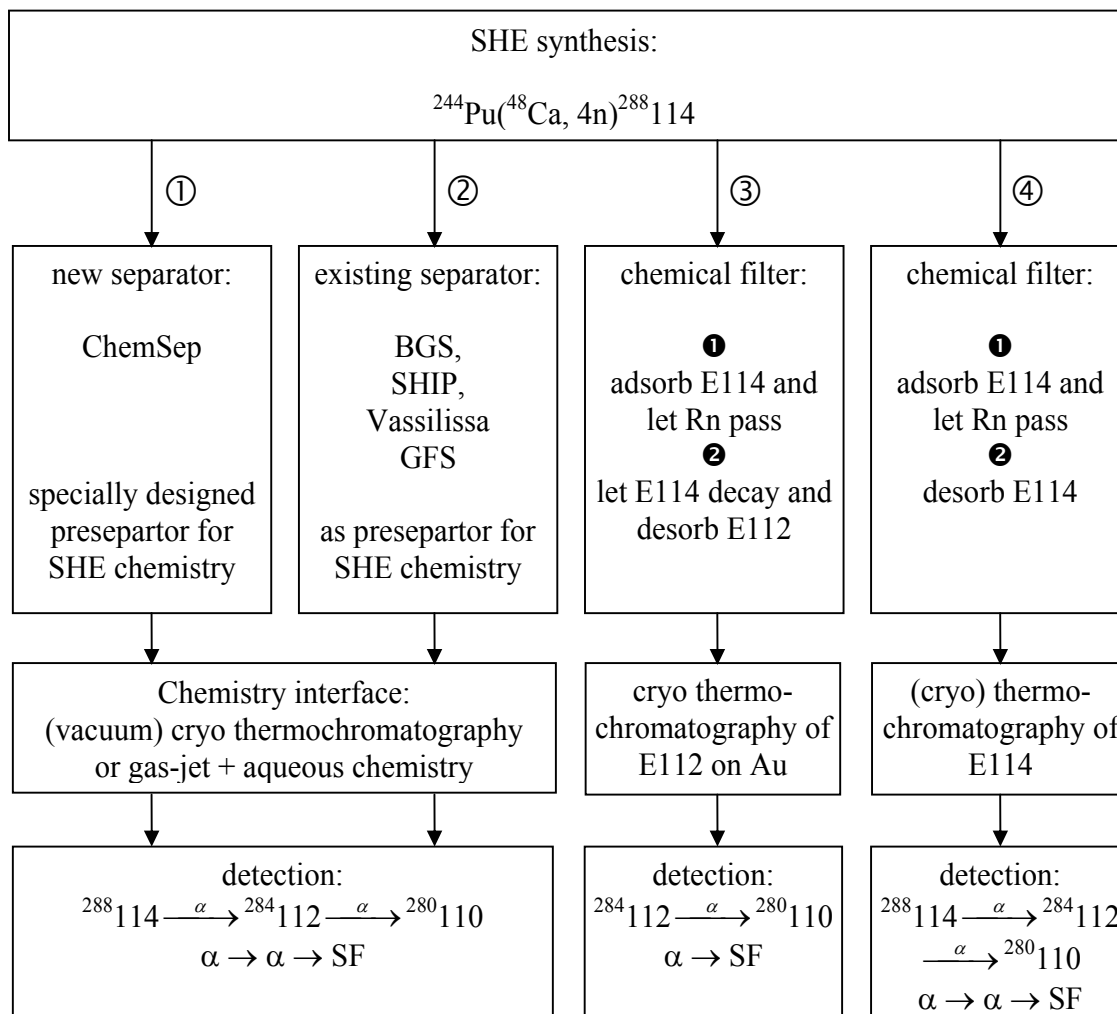


Figure 1. Routes to chemical investigations of SHE providing the unambiguous identification of the investigated nuclides.

Even more challenging than the investigation of elements 112 and 114 is probably the chemical identification of elements 109 through 111, the heavy analogs of Ir, Pt, and Au. Here, no obvious chemical properties or compounds as for instance with group 8 elements are at hand. Again, the use of a kinematic preseparator of nuclear reaction products might considerably facilitate chemical investigations.

The fact that some isotopes of elements 112 and 114 have half-lives of the order of seconds to minutes indicates that there exist similarly long-lived isotopes of elements 113 and 111 that are accessible with current projectile and target combinations. Also here, no chemical separation schemes have been worked out, which would allow an immediate experimental investigation.

To conclude it is fair to say that chemists have made enormous progress in investigating the chemical properties of transactinide elements and certainly the successful hassium experiment will serve as a textbook example for years to come. With the synthesis of relatively long-lived superheavy elements with  $Z=112-118$  chemists are facing an extremely challenging but also rewarding future. The possibility to experimentally characterize the complete 7th row of the periodic table has never become more real than starting with today.

## References

- [1] M. Schädel (Ed.): "The Chemistry of Superheavy Elements", Kluwer Academic Publishers, Dordrecht, 2003.
- [2] Yu.Ts. Oganessian et al., *Eur. Phys. J. A* **5**, 63 (1999).
- [3] Yu.Ts. Oganessian et al., *Nature* **400**, 242 (1999).
- [4] Yu.Ts. Oganessian et al., *Phys. Rev. C* **62**, 041604(R) (2000).
- [5] Yu.Ts. Oganessian et al., *Phys. Rev. C* **63**, 011301(R) (2000).
- [6] Yu.Ts. Oganessian et al., JINR preprint D7-2002-287 (2002).
- [7] R. Eichler et al., *Nature* **407**, 63 (2000).
- [8] P.A. Wilk et al., *Phys. Rev. Lett.* **85**, 2697 (2000).
- [9] Ch.E. Düllmann et al., *Nucl. Instrum. Methods A* **479**, 631 (2002).
- [10] U.W. Kirbach et al., *Nucl. Instrum. Methods A* **484**, 587 (2002).
- [11] M. Schädel, *Acta Physica Polonica B* **34**, 1701 (2002).
- [12] Ch.E. Düllmann et al., *Nature* **418**, 859 (2002).
- [13] A.B. Yakushev et al., *Radiochim. Acta* **89**, 743 (2001).
- [14] A.B. Yakushev et al., *Radiochim. Acta* **91**, 433 (2003).
- [15] R. Eichler et al., contribution to this conference.
- [16] V. Pershina et al., *Chem. Phys. Lett.* **365**, 176 (2002).
- [17] E. Strub et al., *Radiochim. Acta* **88**, 265 (2000).
- [18] Y. Nagame et al., *Czech. J. Phys.* **53**, A299 (2003).
- [19] J.P. Omtvedt et al., *J. Nucl. Radiochem. Sci.* **3**, 121 (2002).
- [20] U.W. Kirbach et al., LBNL Nuclear Science Division Annual Report 1999.
- [21] J.V. Kratz et al., *Radiochim. Acta* **88**, 273 (2000).
- [22] Z.G. Gan et al., *Eur. Phys. J. A* **10**, 21 (2001).